Table S1: Number of observation sites and datasets per subcontinent and region type for PM_1 and OA factor analysis.

	#Sites of PM_1 data	#PM_1 datasets	#Sites of OA factor analysis	#OA datasets
Europe	58	431	46	130
Rural	29	240	25	84
Downwind	17	155	13	24
Urban	12	36	8	22
North-America	35	88	27	68
Rural	17	46	12	35
Downwind	5	7	4	6
Urban	13	35	11	27
Eastern Asia	48	153	45	148
Rural	17	44	17	37
Downwind	11	16	10	16
Urban	20	93	18	95
Latin America & Caribbean	10	18	8	14
Rural	5	7	2	2
Downwind	2	4	3	5
Urban	3	7	3	7
Africa	3	14	3	5
Rural	1	12	1	3
Downwind	1	1	1	1
Urban	1	1	1	1
Southern Asia	5	19	5	19
Rural	1	5	1	5
Downwind	0	0	0	0
Urban	4	14	4	14
South-East Asia & Developing Pacific	3	4	1	2
Rural	2	2	0	0
Downwind	0	0	0	0
Urban	1	2	1	2
Asia-Pacific Developed	7	17	5	12
Rural	5	13	4	10
Downwind	1	1	0	0
Urban	1	3	1	2
Total	169	744	140	398

Table S2: Statistical evaluation of EMAC PM$_1$ components against AMS and ACSM datasets over the tropical and subtropical regions of Latin America and Caribbean, Africa, Southeast Asia and developing Pacific, and Asia Pacific developed during 2000–2020.

Continent	Element	Number of data sets	Mean observed [μgm^{-3}]	Mean predicted [μgm^{-3}]	MAGE [μgm^{-3}]	MB [μgm^{-3}]	NME [%]	NMB [%]	RMSE [μgm^{-3}]
Latin America & Caribbean	OA	21	8.39	5.51	5.46	-2.89	65.04	-34.41	8.12
	SO$_4^{2-}$	19	1.21	0.84	0.44	-0.37	36.29	-30.4	0.59
	NO$_3^-$	19	1.43	0.7	0.91	-0.73	63.78	-50.91	1.93
	NH$_4^+$	19	0.95	0.37	0.58	-0.57	61.46	-60.68	1.11
Africa	OA	14	3.42	5.51	2.41	2.09	70.34	61.04	3.3
	SO$_4^{2-}$	14	1.93	1.23	1.06	-0.71	55.12	-36.55	1.36
	NO$_3^-$	14	0.42	0.8	0.43	0.38	103.78	91.22	0.67
	NH$_4^+$	14	0.8	0.48	0.47	-0.32	58.86	-39.59	0.54
South Asia & Developing Pacific	OA	23	33.26	18.59	19.24	-14.67	57.85	-44.1	28.99
	SO$_4^{2-}$	23	7.01	4.02	4.28	-2.99	61.07	-42.66	5.48
	NO$_3^-$	22	5.39	5.21	3.1	-0.17	57.56	-3.23	4.53
	NH$_4^+$	22	5.24	2.76	3.42	-2.48	65.36	-47.39	5.25
Asia-Pacific Developed	OA	17	5.8	2.54	3.4	-3.26	58.64	-56.14	4.64
	SO$_4^{2-}$	17	4.84	2.3	2.66	-2.53	54.96	-52.35	3.46
	NO$_3^-$	17	0.79	1.53	1.16	0.74	146.03	92.84	1.4
	NH$_4^+$	17	1.58	0.79	0.89	-0.8	56.35	-50.29	1.05

Table S3: Statistical evaluation of EMAC PM$_1$ SOA-sv concentrations against AMS and ACSM datasets over Europe, North America, and Eastern Asia during 2000–2020.

Continent	Region	Number of data sets	Mean observed [μgm^{-3}]	Mean predicted [μgm^{-3}]	MAGE [μgm^{-3}]	MB [μgm^{-3}]	NME [%]	NMB [%]	RMSE [μgm^{-3}]
Europe	all	82	1.18	1.08	0.63	-0.1	53.54	-8.63	0.82
	rural	58	1.02	1.04	0.61	0.02	60.11	2.24	0.82
	DW	13	1.49	0.93	0.81	-0.56	54.14	-37.54	0.95
	urban	11	1.65	1.43	0.52	-0.22	31.44	-13.11	0.68
North-America	all	51	1.89	2.19	1.22	0.3	64.8	15.79	1.69
	rural	28	1.59	1.94	0.96	0.34	60.36	21.62	1.27
	DW	3	4.33	1.98	2.47	-2.35	57.14	-54.34	2.96
	urban	20	1.93	2.56	1.4	0.63	72.5	32.62	1.93
Eastern Asia	all	100	4.69	2.68	2.39	-2.01	50.91	-42.91	3.35
	rural	22	4.03	2.23	2.2	-1.8	54.69	-44.61	2.76
	DW	11	3.84	2.77	1.2	-1.08	31.19	-28.07	1.61
	urban	67	5.05	2.81	2.65	-2.24	52.39	-44.32	3.72

Table S4: Statistical evaluation of EMAC PM$_1$ SOA-lv concentrations against AMS and ACSM datasets over Europe, North America, and Eastern Asia during 2000–2020.

Continent	Region	Number of data sets	Mean observed [$\mu g m^{-3}$]	Mean predicted [$\mu g m^{-3}$]	MAGE [$\mu g m^{-3}$]	MB [$\mu g m^{-3}$]	NME [%]	NMB [%]	RMSE [$\mu g m^{-3}$]
Europe	all	92	1.61	0.7	1.04	-0.91	64.31	-56.57	1.45
	rural	66	1.53	0.67	0.98	-0.86	63.54	-56.24	1.48
	DW	14	1.84	0.59	1.42	-1.25	77.11	-68.15	1.54
	urban	12	1.78	0.99	0.93	-0.78	52.54	-44.13	1.13
North-America	all	60	2.13	1.61	1.21	-0.52	57.04	-24.32	1.48
	rural	33	1.9	1.44	1.12	-0.46	58.77	-24.04	1.36
	DW	4	2.21	1.58	0.67	-0.64	30.34	-28.74	0.88
	urban	23	2.44	1.85	1.45	-0.58	59.33	-23.93	1.7
Eastern Asia	all	109	6.42	0.96	5.46	-5.46	85.06	-85.03	6.81
	rural	27	4.59	0.87	3.72	-3.72	81.19	-81.03	4.61
	DW	11	4.92	1.09	3.83	-3.83	77.87	-77.87	5.11
	urban	71	7.36	0.98	6.38	-6.38	86.72	-86.72	7.69

Table S5: Percentage change (%) in pollutant emissions between the 2000s and 2010s.

	OC	BC	NH$_3$	NO$_x$	SO$_2$	ARO	ALK	OLE	C5H8	TERP	VOC
Southern Asia	27	49	81	112	140	58	93	45	27	129	65
Europe	-9	-45	-3	-38	-55	-47	-44	-41	-52	4	-45
Africa	42	56	60	54	37	61	40	86	76	16	59
South-East Asia & Developing Pacific	30	51	36	75	27	39	43	53	44	99	43
Latin America & Caribbean	53	23	42	41	1	13	12	10	24	127	12
Eurasia	-30	-11	12	-7	-17	-22	-1	-19	10	179	-13
Asia-Pacific Developed	2	-30	23	-28	-14	-11	-29	-30	-28	-13	-12
Middle East	26	50	24	75	93	14	9	7	-3	119	10
North America	-12	-38	19	-47	-65	-36	-45	-59	-73	-4	-42
Eastern Asia	14	48	50	85	44	99	40	10	112	244	58
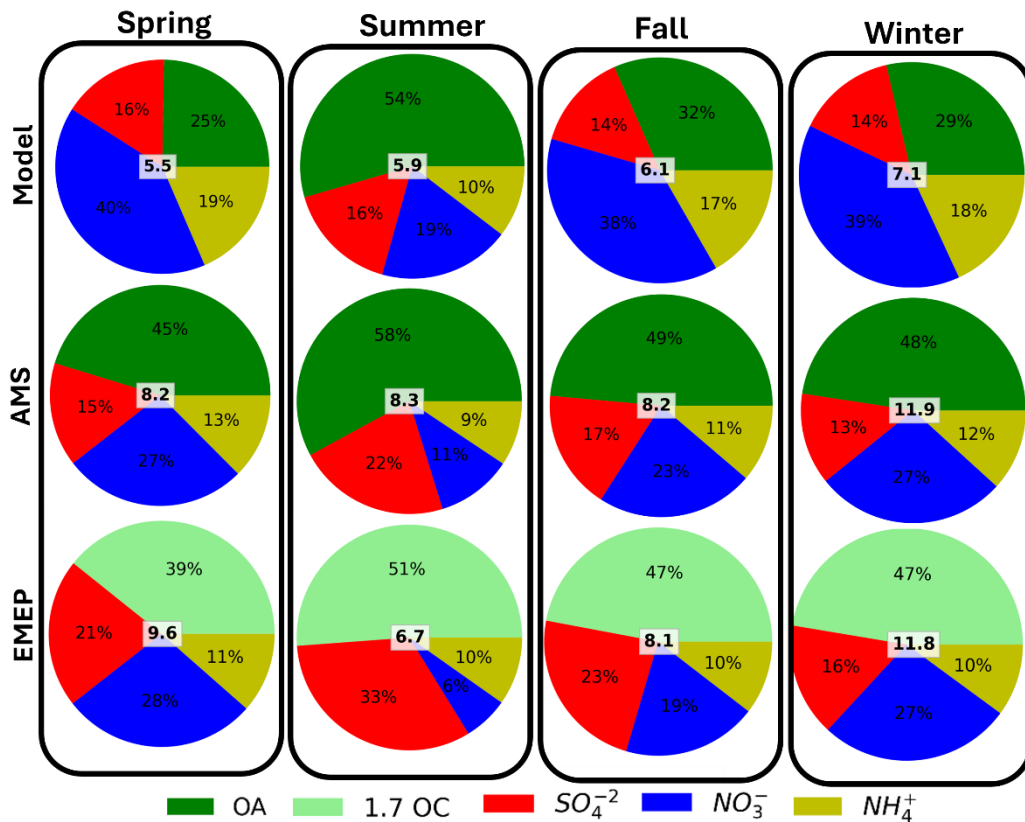
Figure S1. Seasonal aerosol composition in Europe from EMAC simulation, AMS observations and EMEP filter measurements during 2000-2020. The white boxes indicate the seasonal mean of the total PM concentration (in µg m^{-3}), the numbers in the respective pie slices indicate the relative contribution of each element (in %).
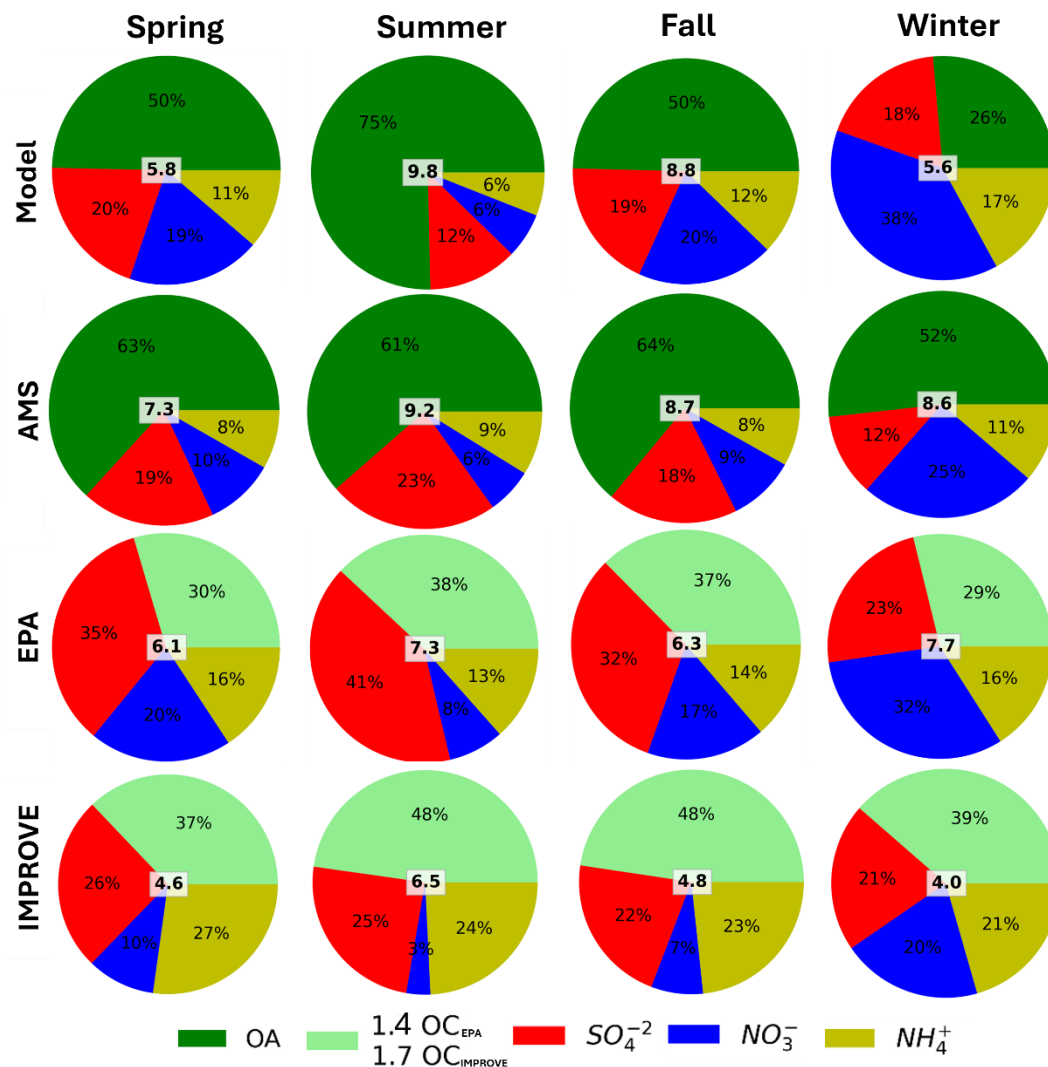
Figure S2. Seasonal aerosol composition in North America from EMAC simulation, AMS observations and IMPROVE and EPA filter measurements during 2000-2020. The white boxes indicate the seasonal mean of the total PM concentration (in µg m^{-3}), the numbers in the respective pie slices indicate the relative contribution of each element (in %).
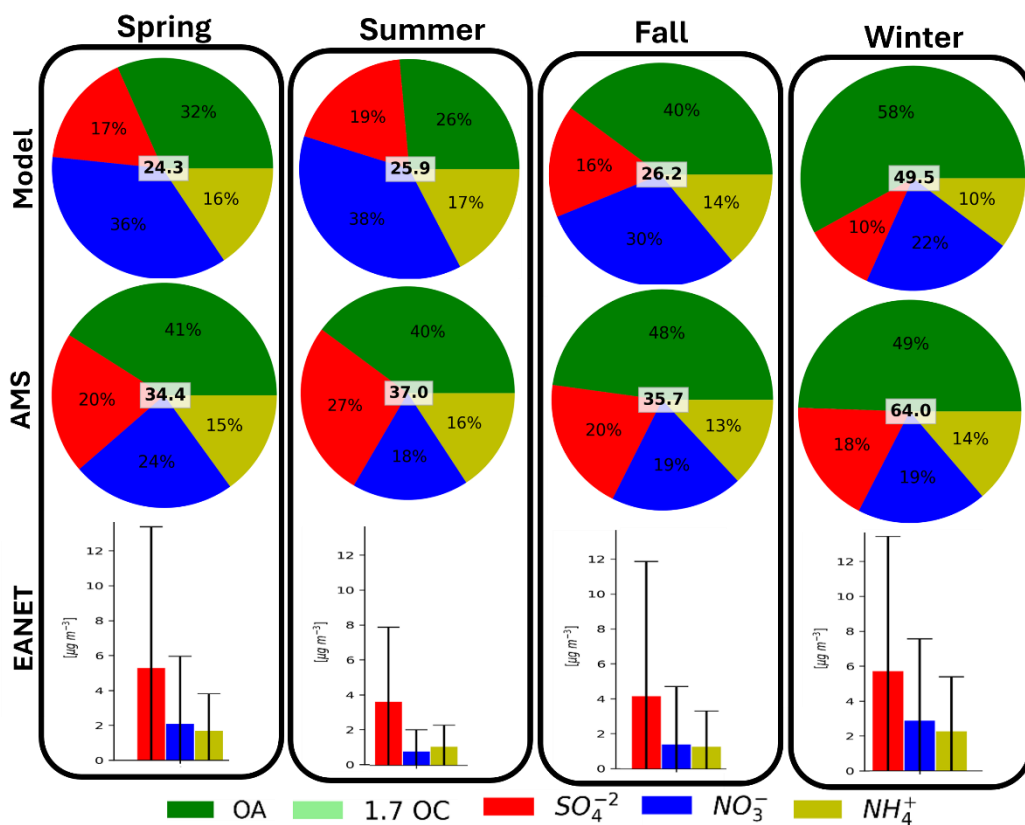
Figure S3. Seasonal aerosol composition in Eastern Asia from EMAC simulation, AMS observations and EANET filter measurements during 2000-2020. The white boxes indicate the seasonal mean of the total PM concentration (in µg m^{-3}), the numbers in the respective pie slices indicate the relative contribution of each element (in %).

100
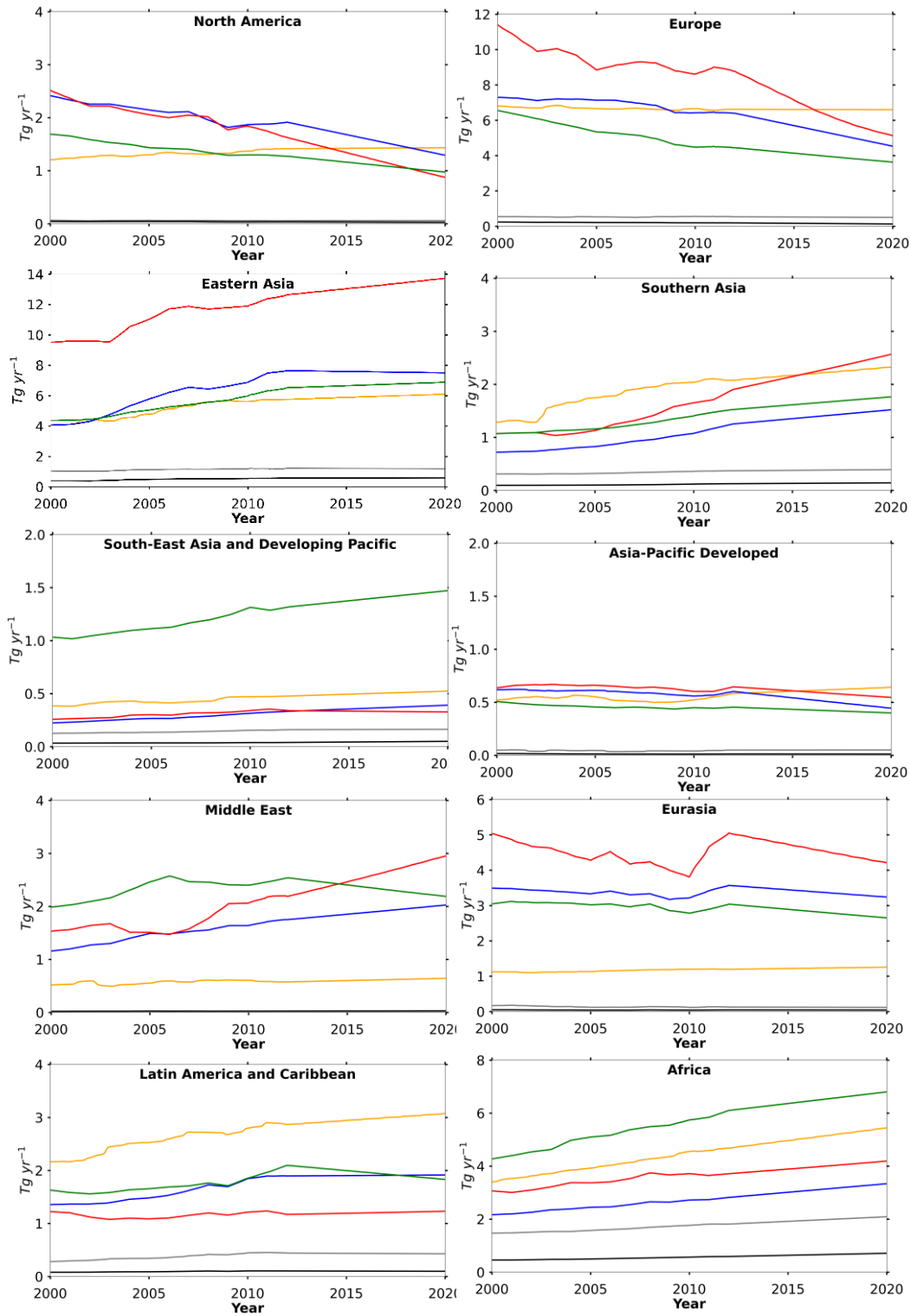
Figure S4. Temporal evolution of pollutant emissions in Tg yr^{-1} of OC (grey), BC (black), NH$_3$ (orange), NO$_x$ (blue), SO$_2$ (red), and VOC (green) during the period 2000-2020 in the 10 regions considered according to WGIII AR6.
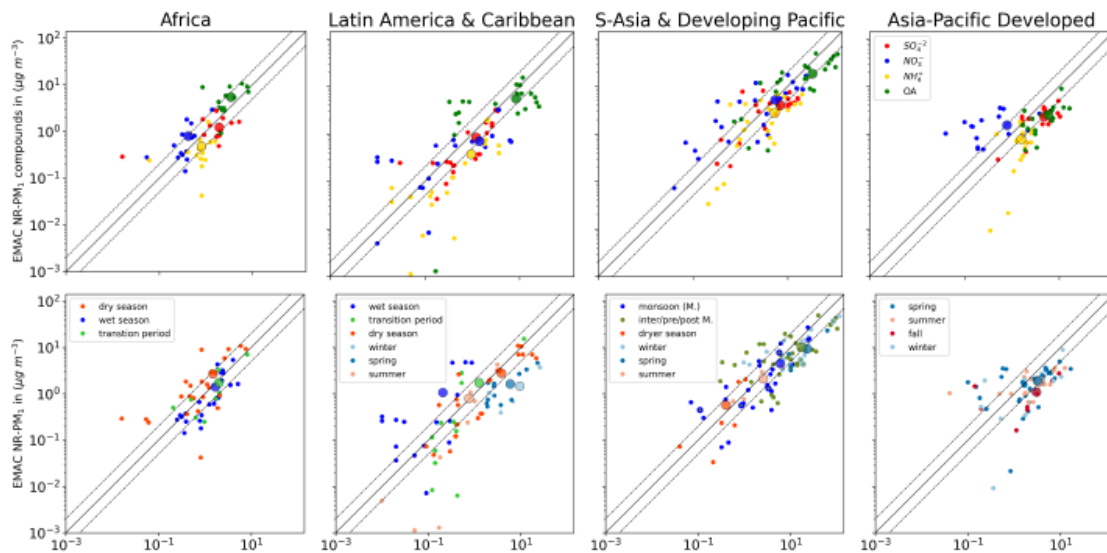
Figure S5. Scatter plots comparing EMAC results for PM$_1$ concentrations (in μg m^{-3}) with AMS and ASCM observations (top row) colored by the season of the field campaign and (bottom row) colored by the chemical composition. Each point represents the mean of the data set. Also shown are the 1:1, 2:1, and 1:2 lines.
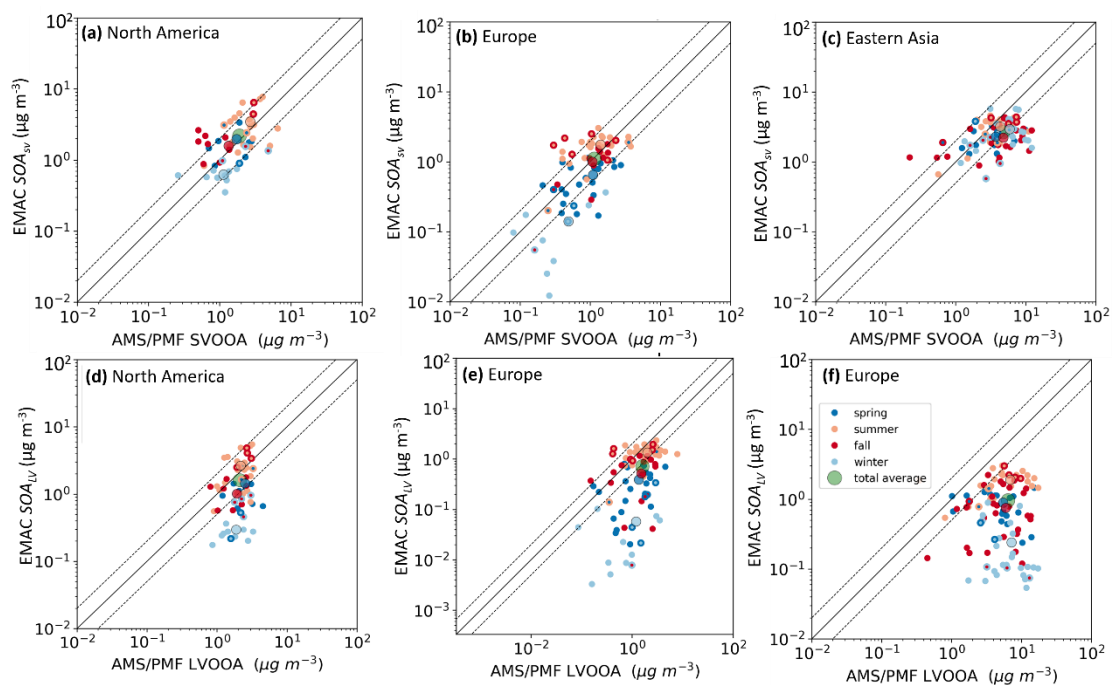
Figure S6: Scatter plots comparing model results for PM_1 SOA_{sv} (a-c) and SOA_{lv} (d-f) concentrations (in μg m^{-3}) with AMS and ASCM observations of M-OOA and L-OOA, respectively, over North America (a, d), Europe (b, e), and Eastern Asia (c, f). Each point represents the data set mean and is colored based on the season of the field campaign. Also shown are the 1:1, 2:1, and 1:2 lines.